

1 **Cause of a lower-tropospheric high-ozone layer in spring over Hanoi**

2 **S. -Y. Ogino¹, K. Miyazaki², M. Fujiwara³, M. I. Nodzu⁴, M. Shiotani⁵, F. Hasebe³, J.**
3 **Matsumoto^{1,4}, J. Witte⁶, A. M. Thompson⁷, H. A. Nguyen-Thi⁸, and T. V. Nguyen⁸**

4 ¹Japan Agency for Marine-Earth Science and Technology, Yokosuka, Japan.

5 ²Jet Propulsion Laboratory/California Institute for Technology, Pasadena, CA, USA

6 ³Faculty of Environmental Earth Science, Hokkaido University, Sapporo, Japan.

7 ⁴Department of Geography, Tokyo Metropolitan University, Hachioji, Japan.

8 ⁵Research Institute for Sustainable Humanosphere, Kyoto University, Kyoto, Japan.

9 ⁶National Center for Atmospheric Research, Earth Observing Laboratory, Colorado, USA.

10 ⁷NASA Goddard Space Flight Center, Maryland, USA.

11 ⁸Aero Meteorological Observatory, Viet Nam Meteorological and Hydrological Administration,
12 Hanoi, Vietnam

13 Corresponding author: Shin-Ya Ogino (ogino-sy@jamstec.go.jp)

14 **Key Points:**

- 15 • Ozonesonde observations in Hanoi, Vietnam, for about fourteen years since 2004 display
16 an ozone enhancement at ~3 km altitude in March–April
- 17 • Enhanced ozone air at ~3 km originates from burning areas west of Hanoi whereas low-
18 ozone near-surface air is marine from the east
- 19 • Model experiments show biomass fires in the Indochina Peninsula are the cause of
20 Hanoi’s enhanced ozone

22 **Abstract**

23 The ozonesonde observations in Hanoi, Vietnam, over fourteen years since 2004 have confirmed
24 the enhancement in lower tropospheric ozone concentration at about 3 km altitude in the spring
25 season. We investigated the evolution of the ozone enhancement from analysis of meteorological
26 data, backward trajectories, and model sensitivity experiments. In spring, air masses over Hanoi
27 exhibit strong height dependence. At 3km, the high-ozone air masses originate from the land area
28 to the west of Hanoi, while low-ozone air masses below about 1.5 km are from the oceanic area
29 to the east. Above 4 km, the air masses are mostly traced back to the farther west area. The
30 chemical transport model simulations revealed that precursor emissions from biomass burning in
31 the inland Indochina Peninsula have the largest contribution to the lower tropospheric ozone
32 enhancement, which is transported upward and eastward and overhangs the clean air intrusion
33 from the ocean to the east of Hanoi. At this height level, the polluted air has the horizontal extent
34 of about 20 degrees in longitude and latitude. The polluted air observed in Hanoi is transported
35 further east and widely spread over the northern Pacific Ocean.

36 **Plain Language Summary**

37 Ozone in the lower atmosphere acts as an air pollutant and is one of the strong greenhouse gases.
38 Understanding chemical and transport processes that control ozone variations provide important
39 implications for the origin of air pollution and for climate change. In the past 20 years, the global
40 tropospheric ozone amount has increased in the tropics and subtropics, with particularly large
41 increases in Southeast Asia. We analyze a seasonal enhancement in ozone over Hanoi, Vietnam,
42 using the 14-year record of upper air ozone measurements that began in 2004. The measurements
43 reveal high levels of ozone particularly during the spring that peak at around 3 km. Numerical
44 model simulations show that agricultural burning in the inland Indochina Peninsula is the major
45 contributor to these high ozone amounts that are transported upward and eastward and overhang
46 the clean air from the ocean to the east of Hanoi. Overtime, the high-ozone air observed in Hanoi
47 is transported further east and widely spread over the northern Pacific Ocean. The identified
48 source of the ozone enhancements and its three-dimensional distribution provide new insights
49 into the impact of agricultural burning over Southeast Asia on large-scale ozone distributions and
50 would benefit global air quality and climate studies.

51 **1 Introduction**

52 Tropospheric ozone is closely related with air quality by producing hydroxyl radicals
53 which control oxidizing capacity of the atmosphere, and is itself an air pollutant and one of the
54 strong greenhouse gases (Brasseur et al., 2003; IPCC, 2013). It is important to describe and
55 understand the mechanism determining the three-dimensional (3D) distribution of ozone and its
56 temporal variation in order to trace the origin of air pollution and to understand climate
57 variability and change.

58 Because of the strong ozone production efficiency, changes in precursor emissions
59 associated with economic and natural activity can lead to substantial changes in ozone in the
60 tropics and subtropics (Cooper et al., 2020; Gaudel et al., 2020; Y. Zhang et al., 2021). While
61 chemistry-climate models predict substantial changes in tropospheric ozone over these regions in
62 the future climate scenarios (e. g., Morgenstern et al., 2017; Revell et al., 2018; Young et al.,
63 2013), the multi model discrepancies remain large in predicted ozone and its radiative forcing
64 (Kuai et al., 2020). Ozone in these regions is also important for human health associated with the

65 high population density and rapid economic development. However, the current in-situ observing
66 network in these regions, for instance from Tropospheric Ozone Assessment Report (TOAR), is
67 clearly insufficient for detailed evaluation of ozone changes for various applications including air
68 pollutant human health impacts (Fleming et al., 2018). Studies using numerical models and
69 limited observations have also illustrated the overall picture of non-local and local atmospheric
70 pollution changes including ozone over South and Southeast Asia, such as wintertime
71 northeasterly transports at low levels and summertime uplifting associated with Asian summer
72 monsoon together with strong impacts of biomass burning (Lawrence & Lelieveld, 2010).
73 Nevertheless, complete 3D structure of ozone in these regions and its driving factors remain
74 unclear mainly due to a lack of detailed observational information. While long-term polar orbit
75 and recent geostationary satellite measurements, such as Geostationary Environment Monitoring
76 Spectrometer (GEMS) (Kim et al., 2020), can provide improved understanding of tropospheric
77 ozone changes, vertical profile information from regular ozonesonde records are essential to
78 study detailed chemical and physical processes.

79 Since September 2004, we have conducted regular ozonesonde observations once or
80 twice a month in Hanoi (21.02°N , 105.80°E), Vietnam, on the Indochina Peninsula (Ogino et al.,
81 2013), as part of the activities of Soundings of Ozone and Water in the Equatorial Region
82 (SOWER; Hasebe et al., 2013) and Southern Hemisphere ADDitional OZonesondes (SHADOZ;
83 Thompson et al., 2012). Hanoi lies at the southeastern edge of the Asian summer monsoon
84 circulation, which is a region of exchange of mass and chemical species between the tropical
85 troposphere and the extratropical stratosphere in the Eastern Hemisphere. Southeast Asia is one
86 of the important regions, where tropospheric ozone shows a long-term increasing trend,
87 suggesting a significant contribution to global ozone change (Gaudel et al., 2018; Y. Zhang et al.,
88 2016). In this regard, chemical and meteorological observations in Hanoi can provide valuable
89 information about regional pollution and transport patterns.

90 Figure 1a shows the seasonal variation in the ozone mixing ratio (OMR) observed in
91 Hanoi averaged over about fourteen years from September 2004 to June 2018. There are two
92 height ranges with a large amplitude of seasonal variation. One is in the upper troposphere and
93 lower stratosphere (UTLS) region (10–20 km), where ozone increases in spring (April to June)
94 and summer (August), and decreases in winter, owing to the seasonal change of transport in the
95 UTLS region. There is a northward transport of low-ozone air in winter due to the Rossby wave
96 response to convective heating over the Maritime Continent and the western Pacific. This is
97 followed by a southward transport of high-ozone air in summer due to the upper-tropospheric
98 anticyclonic monsoon circulation associated with the South Asian High, as shown by Ogino et al.
99 (2013). The other height range for large seasonal variability appears in the lower troposphere,
100 with a peak OMR at around 3 km in the pre-monsoon season (March–April). This study aims to
101 identify the origin of this peak OMR and propose a mechanism for the seasonal ozone
102 enhancement.

103 Figure 2 shows the seasonal variation in the OMR up to 8 km height in the first half of
104 the year, and the atmospheric stability, measured by the Brunt-Väisälä number. It shows two
105 stable layers at ~ 1.5 and ~ 5 km in March, as reported by Nodzu et al. (2006). There is an ozone
106 maximum between those layers that implies a distinct and separate feature.

107 Over the Indochina Peninsula, March is a transition period from the dry season to the
108 rainy season. Intermittent rainfall begins in April (Kiguchi & Matsumoto, 2005; Matsumoto,

109 1997), and the summer monsoon season starts typically in May (He et al., 1987; Hsu et al., 1999;
110 Nguyen-Le et al., 2015; Yanai et al., 1992), the earliest onset in the Asian monsoon region.

111 A similar but smaller ozone increase at the similar height and in the same season was
112 observed in Hong Kong (22.31°N , 114.17°E) (L. Y. Chan et al., 1998; Ogino et al., 2013).
113 Several authors showed that such ozone increases could be attributed to the transport of air
114 polluted by biomass burning over Southeast Asia. Their analysis was based on backward
115 trajectories that tend to pass over the active region of biomass burning, such as the Indian
116 subcontinent and the northern Indochina Peninsula (C. Y. Chan, Chan, Chang, et al., 2003; C. Y.
117 Chan, Chan, Harris, et al., 2003; C. Y. Chan & Chan, 2000; Liao et al., 2021). Liu et al. (2002)
118 further confirmed that Southeast Asian biomass fires are responsible for the ozone enhancement
119 observed in Hong Kong based on model simulations in which biomass burning emissions were
120 eliminated. Recently, Liao et al. (2021) showed that the spring time ozone concentration above
121 Hong Kong has an increasing trend possibly due to the biomass burning increase over the
122 upwind region.

123 Burning activities are maximized in March over the northern part of the Indochina
124 Peninsula (Huang et al., 2016), leading to enhanced surface ozone (Pochanart et al., 2001) and
125 tropospheric column ozone (Sonkaew & Macatangay, 2015) in Thailand, an inland region of the
126 Indochina Peninsula. These studies suggest that air polluted by biomass burning is a plausible
127 cause of the ozone increase at about 3 km above Hanoi in March.

128 Most of the previous studies on the source of the ozone enhancement relied on backward
129 trajectory analyses over Southeast Asia. However, traditional trajectory analysis does not provide
130 any information on which emissions within Southeast Asia contributes most to the ozone
131 enhancement quantitatively. It is also unclear why the ozone increase in March appears at about
132 3 km height above the surface. C. Y. Chan, Chan, Harris, et al. (2003) showed that the increase
133 was accompanied by a stable layer at the bottom of the layer with the ozone increase, and
134 mentioned that the inversion isolated the ozone rich layer from the influence of the local
135 boundary layer. Nonetheless, the formation process and spatial distribution of the stable layer
136 have not yet been fully described, and therefore, its relation to the high ozone layer is not clearly
137 understood. The 3D structure of the ozone enhancement should be described by relating it to
138 meteorological phenomena.

139 In this paper, we carry out a comprehensive study on the ozone enhancement with
140 thermodynamic properties and the detailed analysis of chemical model sensitivity experiments.
141 We first describe the variabilities of the ozone enhancement in March together with background
142 meteorological properties, in which we identify three regimes in terms of ozone variability
143 separated by the stable layers at about 1.5 km and about 4 km.

144 Next, we investigate the source region of ozone precursors that contributes the ozone
145 enhancement above Hanoi by conducting sensitivity experiments using a chemical transport
146 model with the observationally constrained top-down emissions. We further reveal the 3D
147 structure of the ozone enhancement and investigate the mechanisms controlling the ozone
148 enhancement that appears at about 3 km above Hanoi.

149 2 Data and methods

150 We used ozonesonde data collected at the Aero-Meteorological Observatory (21.02°N ,
151 105.80°E), Hanoi, once or twice in each month from September 2004 to June 2018. The

152 observations are described in detail in Ogino et al. (2013). The data were re-processed in 2017
153 and 2018 (Witte et al., 2017, 2018) to obtain the homogenized data (SHADOZ Version 6 data
154 sets). Total ozone from the soundings is in good agreement with satellite overpasses (e.g., from
155 OMI and OMPS; Thompson et al., 2017; Witte et al., 2018) as well as from the Brewer
156 spectrometer co-located at the Aero-Meteorological Observatory launch site. We used
157 operational radiosonde data taken at the Observatory in Hanoi and the Japanese 55-year
158 Reanalysis (JRA-55) objective analysis data (Kobayashi et al., 2015) to investigate
159 meteorological fields.

160 The JRA-55 data were also used in back-trajectory calculations. Isentropic (without
161 considering diabatic vertical motion) back-trajectories were calculated every 30 min using
162 linearly interpolated JRA-55 meteorological reanalysis data (Kobayashi et al., 2015) and the
163 second-order Runge-Kutta method for time integration, as in Hasebe et al. (2007) and Ogino et
164 al. (2013). In each trajectory calculation, an initial point was set at every 200 m height from 100
165 m to the upper troposphere above Hanoi at the ozonesonde launch time in Hanoi.

166 To examine the relative importance of different emission source regions on the ozone
167 enhancement above Hanoi, we performed sensitivity experiments using the global chemical-
168 transport model, CHASER (Sudo et al., 2002) with T42 horizontal resolution (approximately
169 2.8° longitude $\times 2.8^\circ$ latitude) and 32 vertical layers from the surface up to 10 hPa in sigma
170 coordinate. The two-hourly model outputs interpolated onto the constant pressure levels at 1000,
171 990, 970, 930, 870, 790, 700, 610, 530, 460, 400, 350, 300, 260, 230, 200, 176, 153, 133, 116,
172 100, 88, 76, 66, 58, 50, 43, 36, 30, 22, 14, and 10 hPa were used in this study. Note that the
173 updated model, MIROC-Chem (Miyazaki et al., 2017; Watanabe et al., 2011), includes more
174 detailed chemical processes for both troposphere and stratosphere. Nevertheless, CHASER
175 already includes the most important chemical processes in the NOx-CO-Ozone reactions and can
176 be used to evaluate the impact of NOx emissions on ozone productions. In addition, the
177 simulated ozone performance as well as ozone response to NOx emissions are comparable
178 between CHASER and MIROC-Chem (Miyazaki et al., 2020). Thus, the results should not be
179 sensitive to the choice of model.

180 The surface emission of major ozone precursors, such as carbon monoxide (CO), nitrogen
181 oxide (NOx), and nonmethane hydrocarbons, were included in the model based on the published
182 emission inventories (the Emission Database for Global Atmospheric Research (EDGAR)
183 version 4.2 (EC-JRC/PBL, 2011), the monthly Global Fire Emissions Database (GFED) version
184 3.1 (van der Werf et al., 2010), and monthly mean Global Emissions Inventory Activity (GEIA)
185 (Graedel et al., 1993)). We employed daily NOx and CO emissions that were optimized using the
186 assimilation of satellite NO₂ and CO measurements, where the a priori emissions were
187 constructed based upon bottom-up emission inventories (Miyazaki et al., 2015; 2017). These
188 emissions, including both anthropogenic and biomass burning components, used were obtained
189 from the Tropospheric Chemistry Reanalysis version 1 (TCR-1, Miyazaki et al., 2015) and
190 enabled us to evaluate the emission impacts for individual sources.

191 In the sensitivity experiments we eliminated the emissions of ozone precursors from the
192 following three source regions: the Indian subcontinent, the northern Indochina Peninsula, and
193 southern China (Fig.3). We conducted spin-up calculations with the optimized emissions for all
194 regions (i.e., standard emissions) from January 1st to the end of February in each year for 10
195 years from 2005 to 2014. Then, we performed four types of experiments from March 1st to 21st:
196 the control experiment with the standard emissions (Fig. 3), and the three sensitivity experiments

197 with elimination of emission from the above-mentioned three regions, namely the Indian
198 subcontinent, the northern Indochina, the southern China experiments. Because of the non-linear
199 chemistry, the cumulative response from the sensitivity calculations can be different from the
200 total ozone response in the control simulation to some extent as shown by the HTAP modeling
201 works (Turnock et al., 2018; Wild et al., 2012). Nevertheless, they provided important
202 information on relative contributions of emission sources from different regions. The results of
203 the sensitivity experiments will be compared with the control experiment to investigate the
204 relative contributions of individual emission sources to the ozone enhancement over Hanoi. Note
205 that although the period of the model experiment (10 years) is shorter than that of the observation
206 (about 14 years), we consider that the 10-year model data are sufficient for our purpose to
207 describe the climatological feature of the ozone distribution.

208 **3 Characteristics of ozone profiles in March**

209 Figure 4a shows all of the vertical profiles of the OMR observed in Hanoi in March for
210 the whole time period from 2005 to 2018. Figure 4b shows the vertical profiles of the Brunt-
211 Väisälä number for the same ozonesonde observations. The profiles show that the ozone
212 variability is large in three height ranges—0–1.5 km, 1.5–5 km, and 5–8 km—which are
213 separated by two stable layers at ~1.5 and ~5 km first shown by Nodzu et al. (2006). For
214 example, at 1.5–5 km, where the mean OMR in March peaked, the OMR is 50–120 ppbv. At 0–
215 1.5 km the OMR was 0–100 ppbv, and at 5–8 km it is 30–80 ppbv. On the other hand, the
216 variability in OMR is relatively small near the two stable layers at ~1.5 and ~5 km, at 50–80
217 ppbv. The large variability at the three height ranges is confirmed by calculating the standard
218 deviation at each height as shown in Fig. 4c. Figure 4c also shows the frequency distribution of
219 OMR at each height, in which we find different characteristics between the three height ranges.
220 In 1.5–4 km, although the mode is about 60 ppbv, the distribution skews positively and the less-
221 frequent large OMR values (> 80 ppbv) contribute to the ozone enhancement with the large
222 mean value (75–80 ppbv) at this height range. In the layer above 5 km, the distribution shows
223 slight negative skewness, and the mean values are smaller than those in 1.5–4 km, although the
224 modes are similar to or slightly larger than those in 1.5–4.0 km. In the layer below 1.5 km, the
225 frequency distribution skews positively, and both mode and mean decrease as height decrease.

226 Figure 5 shows three typical profiles of the ozone increases at 0–1.5 km (a), 1.5–5 km
227 (b), and 5–8 km (c). On 26 April 2011 (Figure 5a), the OMR increased below the lower stable
228 layer. On 12 March 2007 (Figure 5b), the height profile is similar to that seen in the fourteen-
229 year-averaged height profile of March (Fig. 1b). This case also shows an ozone minimum at 0–
230 1.5 km just below the lower stable layer. Note that there is also a sharp contrast in relative
231 humidity between the levels below and above the lower stable layer: a humid (almost saturated)
232 air mass below, and a relatively dry (about 40% RH) air mass above it. On 27 March 2009
233 (Figure 5c), the OMR increased above the upper stable layer at about 5 km.

234 The ozone increases in each of the three height ranges described above seem to be
235 controlled by different mechanisms. Below, we investigate the mechanism of ozone variation in
236 each layer. First, we investigate whether the ozone density above Hanoi depended on the origin
237 of the air mass by using backward trajectory analysis. Next, we interpret the obtained typical
238 trajectories in terms of transport processes and meteorological conditions.

239 **4 Analysis of a typical case**

240 First, we examine backward trajectories and atmospheric fields for a typical ozone
241 increase at 1.5–5 km and for a typical ozone decrease at 0–1.5 km observed on 12 March 2007
242 shown in Fig. 5b. Figure 6 shows 5-day backward trajectories initialized at 0600 UT (launch
243 time of the ozonesonde) on 12 March 2007 at every 200 m height above Hanoi. The backward
244 trajectories show that the origins of the air masses differed between the height ranges above and
245 below the lower stable layer at about 1.5 km. The air mass in the high-ozone layer at 1.5–4 km
246 can be traced back to the Indochina Peninsula and nearby to the west of Hanoi. The air masses in
247 the low-ozone layer below 1.5 km, on the other hand, originated from the South China Sea and
248 the western Pacific area to the east of Hanoi. This difference is consistent with the observations
249 of humid air below 1.5 km and dry air at 1.5–4 km mentioned in section 3. The change in ozone
250 levels suggests that ozone concentration of the air mass from the Indochina Peninsula was
251 enhanced by biomass burning, while that from the ocean areas was less polluted. The air masses
252 above the upper stable layer (4–6 km) can be traced back to the farther west area than those at
253 1.5–4 km, suggesting that the mechanism that determines the ozone variation in this height range
254 is different from those below about 4 km, and that the long-range transport, and various sources
255 and source regions, such as air pollutions far from Hanoi and stratospheric ozone inputs, can
256 contribute the ozone variations in this height range. In the rest of the paper, we mainly
257 investigate and discuss the ozone variations in the surface-1.5 km and 1.5–4 km height ranges.

258 The pressure and temperature anomaly fields above Hanoi from 7 to 17 March 2007
259 (Figs. 7a, b) reveal a high-pressure, cold-temperature anomaly in the first half of the period. The
260 Brunt-Väisälä number distribution shows clear stable layers at ~1.5 and ~4 km (Fig. 7c).

261 Figure 8a shows the horizontal distribution of the geopotential height and temperature of
262 JRA-55 at 925 hPa on 10 March 2007, 2 days before the ozonesonde observation. The tongue-
263 shaped high-pressure (Fig. 8a, yellow to red), and its associated low-temperature represents an
264 intrusion from eastern China toward Hanoi due to the anti-cyclonic circulation. This structure is
265 characteristic of a cold surge typically seen in winter and spring over Southeast Asia (Compo et
266 al., 1999). The stable layer just above the cold air intrusion was strengthened locally near Hanoi
267 as seen in Fig. 8b and c. Thus, the near-surface easterly flow bringing the low-ozone air mass
268 from the ocean area shown by the backward trajectories in Fig. 6 was associated with this cold
269 surge event, which simultaneously transports the cold air to produce the stable layer at about 850
270 hPa (about 1.5 km). Figure 8c also shows the development of unstable layer from the surface to
271 about 600 hPa (4–5 km) above the land regions of the Indian Subcontinent and the Indochina
272 Peninsula. This is likely caused by the development of a deep mixed boundary layer (0600UT
273 corresponds to about noon time near the longitude region concerned) generated by the increased
274 heating over the land surface through the seasonal transition from winter to summer (Nodzu et al.,
275 2006; Ogino et al., 2010). This suggests that the polluted high-ozone air over the Indian
276 Subcontinent and Indochina Peninsula can be lifted to 4–5 km level and be transported eastward
277 to the ozone enhancement above Hanoi.

278 **5 Model experiment**

279 Here we use the chemical transport model simulations to further explore the relative
280 contributions of individual emission source regions (c.f., Section 2). Figure 9 shows a time-
281 height section of the 10-year mean of monthly mean OMR at the grid point near Hanoi in the
282 control experiment using the standard optimized emissions constrained by satellite observations.

The control experiments well reproduce the seasonal variations in OMR. For instance, the simulated OMR reproduces the observed relative ozone maxima in March at about 700 hPa (about 3 km in height, Fig. 2), although the simulated peak value (65 ppbv) is smaller than the ozonesonde measured peak (85 ppbv) probably due to the coarser vertical resolution of the model and the model biases, such as the low stratospheric ozone input and the insufficient chemical production of ozone as suggested by Park et al. (2021). The model also well reproduces the observed relative timing and location of the seasonal and vertical distributions, such as the two maxima in May-June and September-October in the upper troposphere (400–200 hPa). About a one-month delay in the simulated seasonal march relative to the observed climatological variations in the upper troposphere implies systematic biases in the model chemical and physical processes, but it does not appear in the lower troposphere.

Temporal variability in ozone reproduced in the model shows similar features to the observation shown in Fig. 4. Figures 10a and 10c show that the ozone variability is large at three height ranges — 950–900 hPa (near-surface), 790–700 hPa (about 3 km, where the mean ozone maximum exists), and above 500 hPa — which are separated by the two stable layers at about 900 hPa and at 650–400 hPa. Frequency distribution (Fig. 10c) shows positive skewness at 790–700 hPa, and the outlying large ozone values contribute to the mean value at this pressure level, which is the same feature with the observational results shown in Fig. 4. Thus, the characteristic of ozone variability observed by the once- or twice-monthly ozonesondes was appropriately reproduced by the model experiment; conversely the ozonesonde observation has sampled ozone profiles without strong sampling biases.

At 700 hPa, the control experiment (Fig. 11a) reveals high OMR (> 62 ppbv) over each of the eastern parts of the two regions: the northeastern part of Indian subcontinent and northeastern part of the Indochina Peninsula. By eliminating the ozone precursor emission over the Indian subcontinent region, the ozone enhancement over northeast India disappears, whereas the one over the northeast Indochina Peninsula remains (Fig. 11b). The ozone enhancement over the northeastern part of the Indochina Peninsula, including the Hanoi location, was associated with emissions over the northern Indochina region (Fig. 11c). In the case of the southern China experiment, the Indochina signal remains (Fig. 11d). The contribution from emissions over each region can be more clearly seen by subtracting the result of each sensitivity experiment from that of the control experiment, as seen in Fig. 11e–g. We find that the Indochina emission produces the ozone enhancement above Hanoi and its impact at 700 hPa (about 3 km) spreads around Hanoi with horizontal extent of about 20 degrees in latitude and longitude (Fig. 11f).

In terms of vertical structure, the ozone enhancement above Hanoi originates from the northern Indochina emission (Fig. 12c, f). Figure 12f shows that the ozone enhancement over Indochina (93°E – 106°E) is tilting and overhanging toward the east. This is the morphological reason why the ozone enhancement above Hanoi appears at about 3 km height above the surface. Thus, the overhanging ozone enhancement is formed by the polluted high-ozone air transport from the inland Indochina Peninsula over the clean oceanic air intrusion from the east near the surface. This picture is consistent with the backward trajectories for the typical case shown in the previous section (Fig. 6) and the zonal wind distribution shown in Fig. 13. Meanwhile, the distribution of carbon monoxide (CO) (Fig 14) almost coincides with that of ozone. Because of its direct emissions and relatively long lifetime in the atmosphere, enhanced CO in this region can be regarded as an indicator of air originated from biomass burning and less affected by stratospheric sources unlike ozone. The simultaneous enhancement of ozone and CO indicates

328 that the ozone enhancement occurs due to the biomass burning associated with slash-and-burn
329 agriculture and agro-residue burning.

330 Figure 15a shows the vertical profiles of OMR above Hanoi obtained in the control and
331 sensitivity experiments. The ozone enhancement appears at 700 hPa with the value of 66 ppbv in
332 the control experiment. The OMR at the peak altitude (700 hPa) reduces to 63, 44, and 64 ppbv
333 in the Indian Subcontinent, the northern Indochina, and the southern China experiments,
334 respectively. The contributions of the ozone precursors emitted over the Indian Subcontinent, the
335 northern Indochina, and the southern China regions on ozone at 700 hPa over Hanoi were
336 estimated from the sensitivity experiments to be 3 ppbv (the Indian Subcontinent experiment), 21
337 ppbv (the northern Indochina experiment), and 2 ppbv (the southern China experiment) (Fig.
338 15b), accounting for 5% (the Indian Subcontinent experiment), 33% (the northern Indochina
339 experiment), and 3% (the southern China experiment) changes (Fig. 15c), with additional 59%
340 contributions from other regions. Although these values from the sensitivity experiment results
341 involve ambiguity to some extent owing to the non-linear chemistry as mentioned in Section 2,
342 they clearly highlight the dominant contribution from the northern Indochina region to the lower
343 tropospheric ozone enhancement above Hanoi in spring.

344 Near the surface below the lower stable layer at about 1.5 km, the contribution from the
345 southern China precursors is predominant (account for about 20% of ozone production), with
346 almost negligible contributions from the Indian Subcontinent and the northern Indochina
347 precursors (Fig. 15). The winter monsoon northeasterly could play a role in bringing the polluted
348 air from the southern China region. This may seem inconsistent with the explanation that the
349 intrusion of less polluted, oceanic air masses associated with the winter monsoon flow is
350 responsible for the low mean ozone concentration the surface as mentioned in Section 4, but it
351 can be reasonably interpreted as follows. Although the mean flow in March is almost easterly,
352 the daily wind can fluctuate depending on the short time scale disturbances, such as cold surges,
353 which resulted in the mixture of the dominant transport of the clean oceanic air from the east and
354 of the intermittent intrusion of the polluted air from the south China region to the north and the
355 northeast of Hanoi. Such an intermittent intrusion of high ozone event associated with the
356 monsoon activities is a new insight for the near-surface ozone variation, details of which should
357 be studied in a further investigation.

358 Finally, let us look at the global spread of ozone produced by the Northern Indochina
359 emission. Figure 16 shows the ozone amount generated from the northern Indochina emission
360 during March 1–20 in the middle troposphere (610 hPa). The chemical transport model suggests
361 that ozone originating from northern Indochina can be transported widely eastward in the middle
362 troposphere across the Pacific Ocean, and reach the North America and the North Atlantic Ocean,
363 while the signals are also accumulated over the southern mid-latitudes in the middle troposphere.
364 This feature has been reported by several studies based on the airplane experiments, such as
365 Pacific Exploratory Mission-West (PEM-West) (Hoell et al., 1997) and Transport and Chemical
366 Evolution Over the Pacific (TRACE-P) (Zhang et al., 2003). The present study has revealed the
367 detailed and 3D structure near the source region and also over remote regions, which would
368 provide important insights into the impact of biomass burning over Southeast Asia on global
369 ozone distributions and radiation balance of the atmosphere. Further studies using the presented
370 model simulations along with detailed validation using ozonesonde measurements would benefit
371 global air quality and chemistry-climate studies, for instance, under the Hemispheric Transport
372 of Air Pollution (HTAP) project.

373 **6 Discussion**

374 On the basis of the analyses of backward trajectories, meteorological fields, and the
375 numerical experiments, we propose a mechanism, illustrated in Fig. 17, for the increase of ozone
376 frequently observed at ~3 km above Hanoi in March. During this month, the atmosphere is
377 relatively unstable, allowing shallow convection to develop below the upper stable layer at ~5
378 km over the inland Indochina Peninsula and the Indian Subcontinent. It is well known that the
379 occurrence of biomass burning in these regions increases during February to April (Huang et al.,
380 2016). The near-surface polluted air is well mixed by shallow convection up to the height of the
381 upper stable layer. This well-mixed air is advected by the westerly flow; but the air below the
382 lower stable layer is blocked by the near-surface easterly flow with the oceanic, less-polluted
383 airmass, and only the air above the lower stable layer can reach the eastern region of the
384 Indochina Peninsula. The main source of the enhanced ozone above Hanoi originates from the
385 ozone precursors emitted over the northern part of the Indochina Peninsula. The polluted high-
386 ozone air at the peak height covers the region with a horizontal scale of about 20 degrees in
387 longitude and latitude over the northeastern part of the Indochina Peninsula around Hanoi. The
388 airmasses are transported further eastward and upward and spread over the northern Pacific
389 Ocean in the middle troposphere.

390 This mechanism occurs from March to April, because land heating is not strong enough
391 in January and shallow convection does not reach 4–5 km as a result (Nodzu et al., 2006). Also it
392 is before the rainy season that starts in May over the Indochina Peninsula (e.g., He et al., 1987;
393 Hsu et al., 1999; Yanai et al., 1992). Thus, the ozone transport and distribution from March to
394 April are characterized not only by the seasonality of biomass burning, but also by the monsoon
395 transition from the dry season to the rainy season.

396 Our results show that the ozone increase at ~3 km in March above Hanoi resembles that
397 seen over Hong Kong. A polluted air mass from the same origin may sometimes pass over both
398 Hanoi and Hong Kong. Such polluted air masses are considered to originate from the north of the
399 Indochina Peninsula, which is closer to Hanoi than to Hong Kong. This is consistent with the fact
400 that the climatological signal of the ozone increase is clearer in Hanoi than in Hong Kong as seen
401 in Fig. 1 of Ogino et al. (2013).

402 We showed that a stable layer (at ~5 km) exists at the top of the ozone increase, adding to
403 the finding of the stable layer at the bottom of the ozone increase by L. Y. Chan et al. (2000) and
404 C. Y. Chan et al. (2003). The ozone increase appears between two stable layers at ~1.5 and ~5
405 km. The ozone density above the upper stable layer seems to be separated from that below it and
406 is controlled by a different mechanism. L. Y. Chan et al. (2000) and C. Y. Chan et al. (2003)
407 stated that the lower stable layer isolates the layer with the ozone increase at ~3 km from the
408 influence of the local boundary layer. We add that the horizontal advection of clean, oceanic air
409 associated with the cold surge passage is essential for the isolation.

410 Recent studies suggested that stratospheric ozone intrusion plays an important role in
411 tropospheric ozone variations including the spring ozone enhancement in spring above Hong
412 Kong (Liao et al., 2021; Zhao, Hu, et al., 2021; Zhao, Huang, et al., 2021). Although we cannot
413 discuss quantitatively the stratospheric contribution to the spring ozone enhancement above
414 Hanoi, because the experiment in this study tested the sensitivity to the surface emissions of
415 ozone precursors, a certain amount of stratospheric ozone can contribute to the spring ozone
416 enhancement above Hanoi. However, if we look at the spatial distribution as seen in Figs. 12 and

417 13, the observed ozone enhancement in the lower troposphere above Hanoi is clearly connected
418 with the surface emission over the Indochina Peninsula. In any case, the geographical difference
419 between Hong Kong and Hanoi can lead to different stratospheric contributions. The 3D
420 transport process should be clarified in more detail in future investigations to understand the
421 relative importance of stratospheric intrusion.

422 **7 Conclusions**

423 Regular ozonesonde observations for about fourteen years above Hanoi revealed an
424 ozone increase at ~3 km in March. The ozone densities in March showed large temporal
425 variability at three height ranges that are separated by two stable layers at ~1.5 and ~5 km.
426 Meteorological and backward trajectory analyses showed that a typical ozone enhancement at ~3
427 km originated from air polluted over the Indochina Peninsula and a typical decrease below 1.5
428 km was caused by the advection of the clean oceanic air associated with a cold surge event.

429 We conducted a sensitivity experiment using a chemical transport model, in which we
430 eliminated the emissions of ozone precursors (CO and NO₂) in three regions (the Indian
431 Subcontinent, the northern Indochina, and southern China). The ozone concentration above
432 Hanoi was most effectively suppressed when emissions of ozone precursors over northern
433 Indochina Peninsula are eliminated. This suggests that air pollution from northern Thailand
434 (probably due to biomass burning) contributed most to the ozone increase in Hanoi. The model
435 showed that the ozone increase originated from the northern Indochina Peninsula emission
436 expanded in the lower troposphere over an area of about 20 degrees in longitude and latitude.
437 The model also reproduced the ozone decrease near the surface due to the clean air intrusion
438 associated with the winter monsoon easterly wind. The polluted air was further transported
439 eastward into the middle and upper troposphere across the Pacific Ocean, and some of it reached
440 the west coast of the USA.

441 We propose that the ozone increase at 3 km over Hanoi in March was caused by the
442 eastward advection of polluted, high-ozone air that was well mixed up to the stable layer at ~5
443 km over the land mass of the Indochina Peninsula to the west of Hanoi, and by the westward
444 advection of less-polluted, low-ozone air from the oceanic area to the east of Hanoi associated
445 with a cold surge event below the stable layer at ~1.5 km. Such a mechanism occurs only
446 between March and April, after the development of active shallow convection and before the
447 start of summer monsoon rainfall. We conclude that the ozone enhancement over Southeast Asia
448 is caused not only by the biomass burning enhancement, but also by atmospheric circulation
449 system formed in pre-monsoon season. The circulation system determines the detailed 3D
450 structure of ozone distribution.

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459 **Data Availability Statement**

460 The ozonesonde data used in this study are available in the SHADOZ archive
461 (<https://tropo.gsfc.nasa.gov/shadoz/>). JRA-55 data can be obtained from the Japan
462 Meteorological Agency (http://jra.kishou.go.jp/JRA-55/index_en.html). The radiosonde data at
463 the Hanoi station are available in the archive site of University of Wyoming
464 (<http://weather.uwyo.edu/upperair/sounding.html>). The data of the numerical experiment
465 presented in Sec. 5 are currently available from a Dropbox folder
466 https://www.dropbox.com/sh/prlpfracqsa1c1a/AABVkwuT_QTkTW2AmJ9PcCBda?dl=0 and
467 will be permanently stored in the data base of Japan Agency for Marine-Earth Science and
468 Technology, <http://ebcrpa.jamstec.go.jp/tcr2>.

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- 677
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679 **Figure captions**

680 **Figure 1.** (a) Fourteen-year-averaged seasonal variation in ozone mixing ratio (OMR) obtained
681 by calculating monthly-mean values at each height to 20 km from September 2004 to June 2018.
682 Each monthly-mean value is plotted at each vertical dotted line. (b) Vertical profiles of annual-
683 mean OMR (dashed line), standard deviation of fourteen-year-averaged seasonal variation in
684 OMR (thin solid line), and standard deviation normalized by the annual-mean OMR at each
685 height (thick solid line). These panels update Fig. 1 in Ogino et al. (2013) by adding seven years
686 data.

687 **Figure 2.** Fourteen-year-averaged seasonal variation in ozone mixing ratio obtained by
688 calculating monthly-mean values at each height to 8 km from January to June from 2004 to
689 2018. Black contours show fourteen-year-averaged seasonal variation of the Brunt-Väisälä
690 number ($\text{rad}^2 \text{s}^{-2} \times 10^4$). Each monthly-mean value is plotted at each vertical dotted line.

691 **Figure 3.** The rectangles show the regions where the ozone precursor emissions were eliminated
692 in the sensitivity experiments: (Blue) the Indian Subcontinent, (Green) the northern Indochina
693 Peninsula, and (Red) southern China. The gray shade shows the \circ carbon monoxide and (b)
694 nitrogen oxide emissions from the surface in March averaged for 10 years from 2005 to 2014
695 adopted in the control run of the numerical experiment. A cross mark shows the location of
696 Hanoi, Vietnam.

697 **Figure 4.** Vertical profiles of (a) ozone mixing ratio and (b) Brunt-Väisälä number observed by
698 the ozonesondes in March for the whole time period (2005–2018). Blue line shows the mean of
699 all data in each panel. (c) Frequency distribution (shade), the mean (white solid line) and one
700 standard deviation on the either side of the mean (white dashed line) of ozone mixing ratio.

701 **Figure 5.** Vertical profiles of the ozone mixing ratio (red), the Brunt-Väisälä number (blue), and
702 relative humidity (black) on (a) April 26, 2011, (b) March 12, 2007, and (c) March 27, 2009.

703 **Figure 6.** Backward trajectories initialized at 0600 UT on 12 March 2007 plotted on (a)
704 longitude-height section and (b) longitude-latitude section. Color of each trajectory differs
705 depending on its starting height as found in \circ .

706 **Figure 7.** Time-height sections of (a) pressure anomaly, (b) temperature anomaly, and (c) Brunt-
707 Väisälä number (N^2) drawn from operational radiosonde data collected in Hanoi from 7 to 17
708 March 2007. The anomalies are deviations from the 10-day mean at each height. Vertical solid
709 line denotes the ozonesonde observation at 0600 UT on 12 March 2007.

710 **Figure 8.** Horizontal distributions of (a) geopotential height (colors) and temperature (contours)
711 at 925 hPa, (b) vertical stability (vertical gradient of potential temperature $-\partial\theta/\partial p$) at 862.5 hPa,
712 and (c) longitude-pressure section of vertical stability at 20°N at 0600 UT on 10 March 2007
713 drawn with JRA-55 data. The regions under the ground surface are masked with white color.
714 Black cross marks in \circ and \circ denote the location of Hanoi. A white vertical line in \circ denotes the
715 longitude of Hanoi.

716 **Figure 9.** Time-height cross section of the 10-year mean of monthly mean OMR at the grid near
717 Hanoi reproduced in the control experiment of the sensitivity experiments using the chemical

718 transport model, CHASER. Each monthly-mean value is plotted at each vertical dotted line. The
719 regions under the ground surface are masked with white color.

720 **Figure 10.** Vertical profiles of (a) ozone mixing ratio and (b) vertical gradient of potential
721 temperature reproduced in the control experiment. All the 2-hourly profiles in March 1–21 for 10
722 years from 2005 to 2014 are plotted in grey lines, and the mean of each parameter is plotted in
723 blue line in each panel. (c) Frequency distribution (shade), the mean (white solid line) and one
724 standard deviation on the either side of the mean (white dashed line) of ozone mixing ratio.

725 **Figure 11.** The mean horizontal ozone distributions at 700 hPa for the period from March 1st to
726 20th obtained from the 10-year sensitivity model experiments. (Upper panels) Ozone mixing
727 ratio for (a) the control, (b) the Indian subcontinent, (c) the northern Indochina, and (d) the
728 southern China experiments. (Lower panels) The ozone production due to the ozone precursors
729 emitted from (e) the Indian subcontinent, (f) the northern Indochina, and (g) the southern China
730 regions. A cross mark in each panel shows the location of Hanoi. The regions under the ground
731 surface are masked with white color.

732 **Figure 12.** Same as Fig. 11 but for zonal-vertical sections at 20.9°N. Contours show the vertical
733 stability estimated by vertical gradient of potential temperature in unit of 10^2 K/hPa. A white
734 vertical line in each panel shows the longitude of Hanoi.

735 **Figure 13.** Zonal-vertical section of the zonal wind at 20.9°N reproduced in the control
736 experiment. Contours show the vertical stability estimated by the vertical gradient of potential
737 temperature in unit of 10^2 K/hPa. A white vertical line shows the longitude of Hanoi. The regions
738 under the ground surface are masked with white color.

739 **Figure 14.** As in Fig. 13, but for the CO mixing ratio in the control experiment.

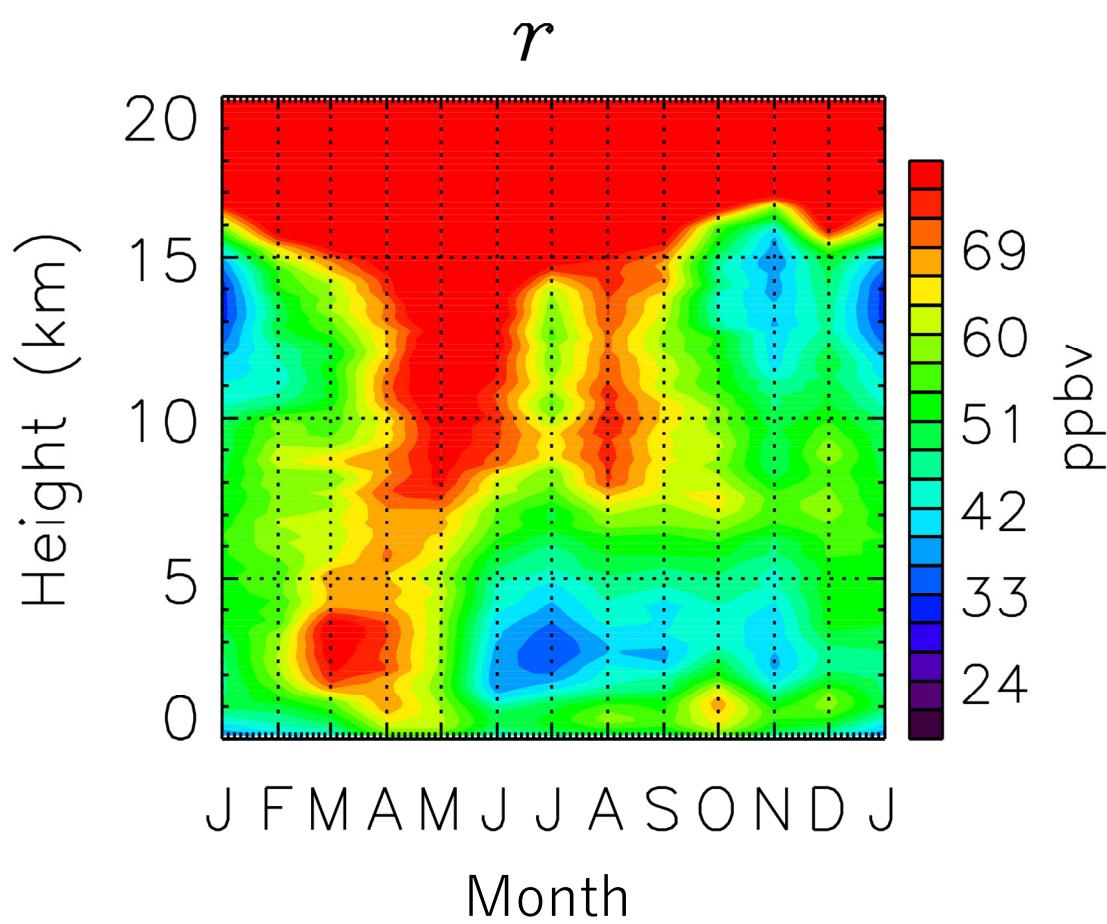
740 **Figure 15.** Vertical profiles of (a) ozone mixing ratio obtained from the sensitivity experiment
741 over the model gridpoint nearest to Hanoi. Each line represents a result from (black) the control,
742 (blue) the Indian Subcontinent, (green) the Northern Indochina, and (red) the Southern China
743 experiment. (b) Its difference between the control experiment and each of the sensitivity
744 experiment, which means ozone mixing ratio contribution from each emission region, and (c)
745 contribution percentage due to the ozone precursor emission in each sensitivity experiment
746 (obtained by dividing the difference values show in (b) by the amount obtained from the control
747 experiment shown by the black line in (a)) are also shown.

748 **Figure 16.** Global distribution of ozone mixing ratio at 610 hPa obtained from the difference
749 between the control experiment and the northern Indochina experiment. A cross mark denotes
750 the location of Hanoi.

751 **Figure 17.** Schematic diagram of the proposed mechanism for the ozone increase at 3 km above
752 Hanoi in March.

Figure 1.

(a)



(b)

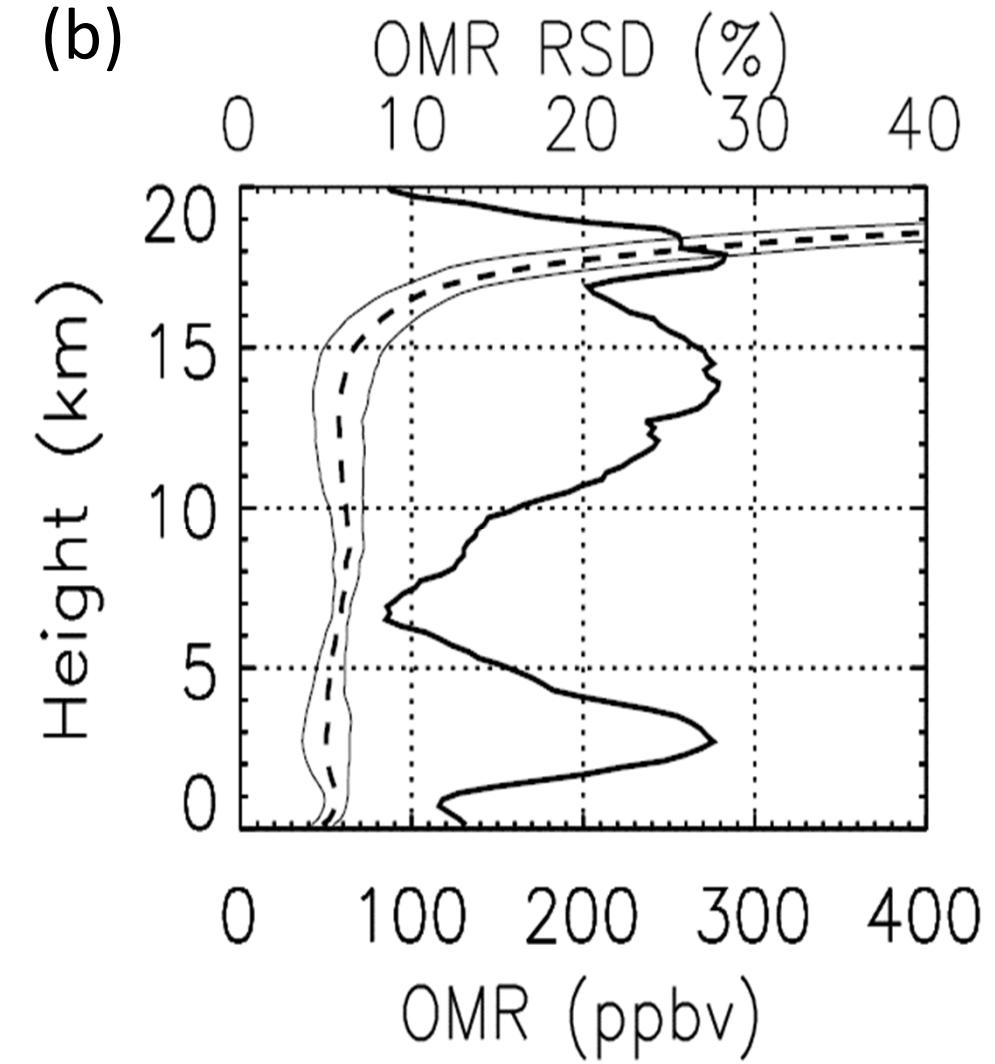


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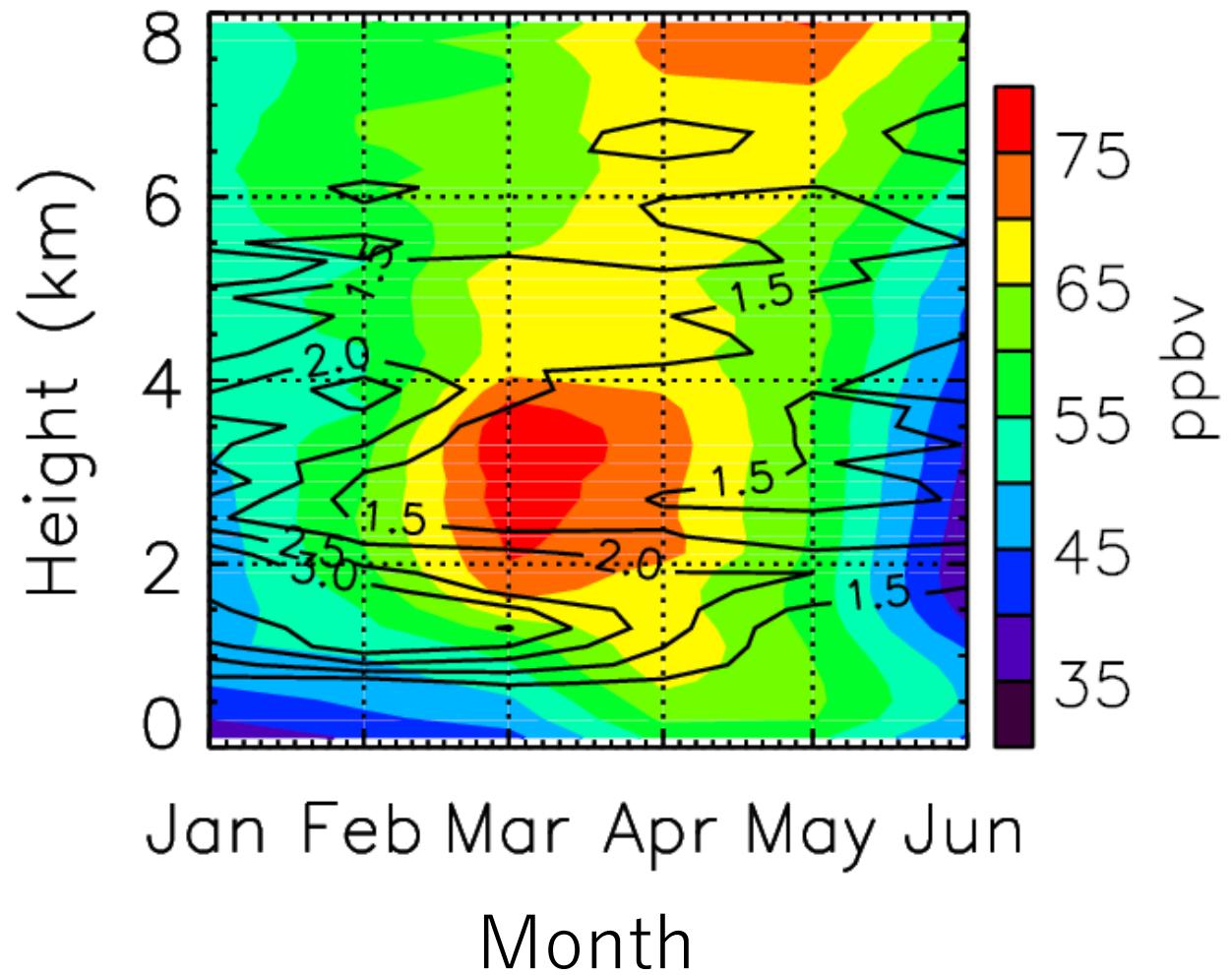


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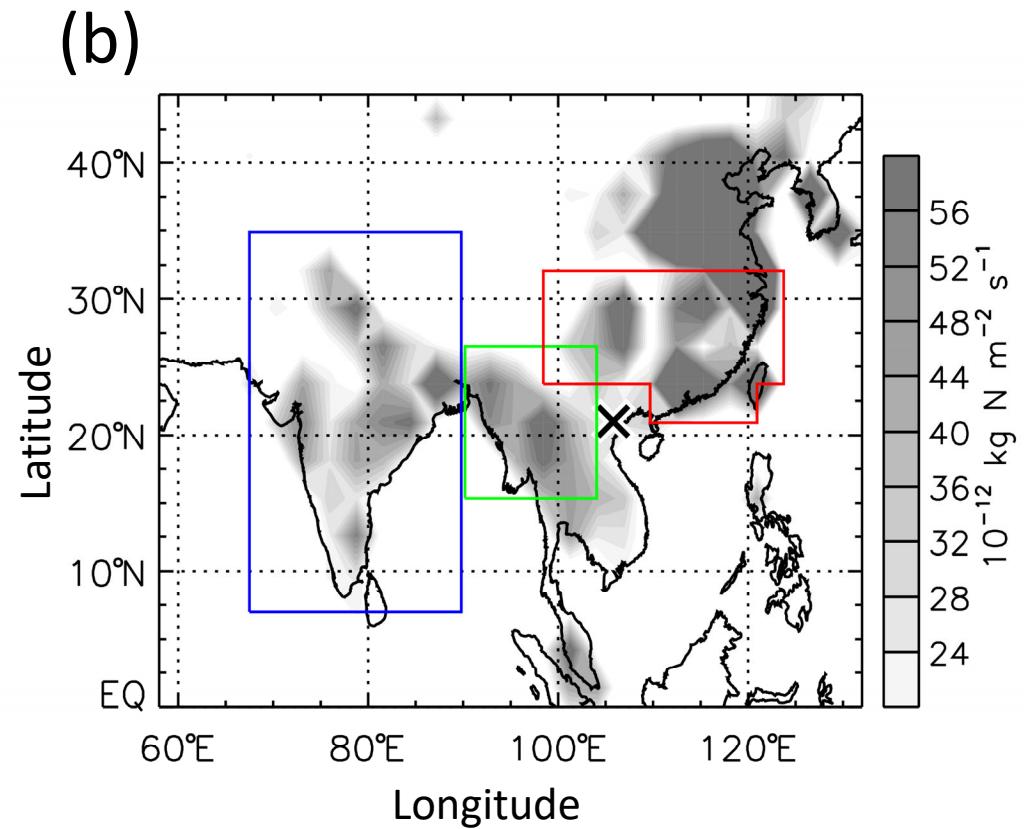
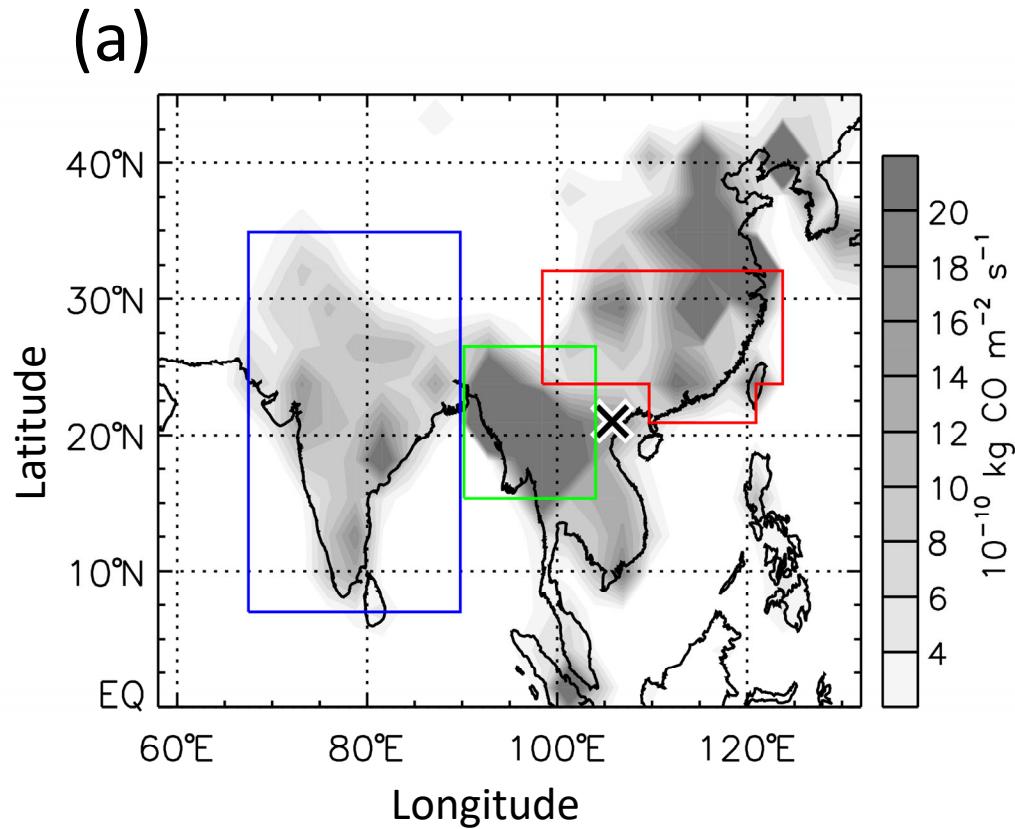


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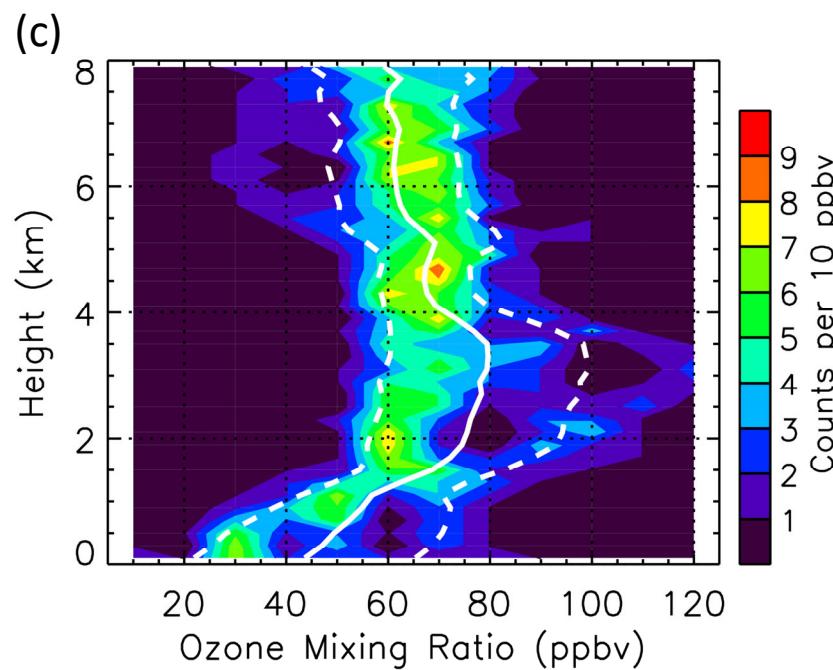
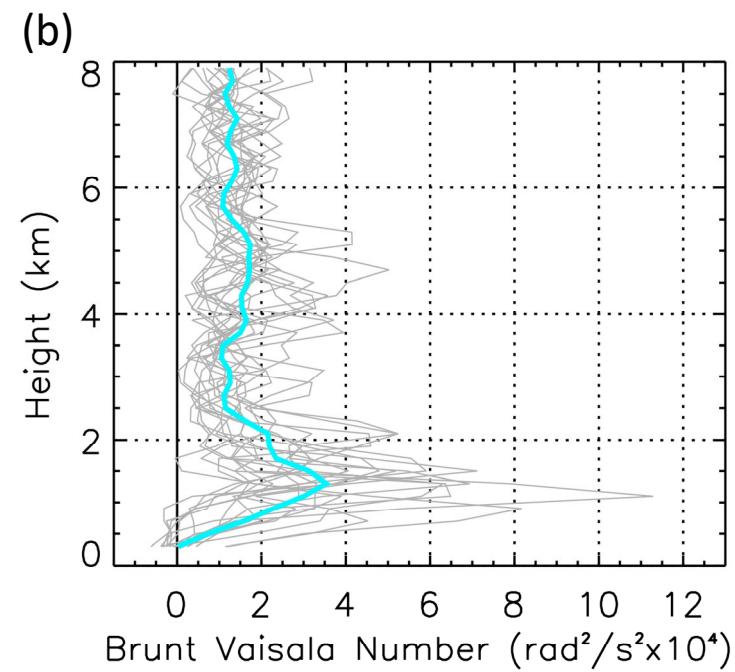
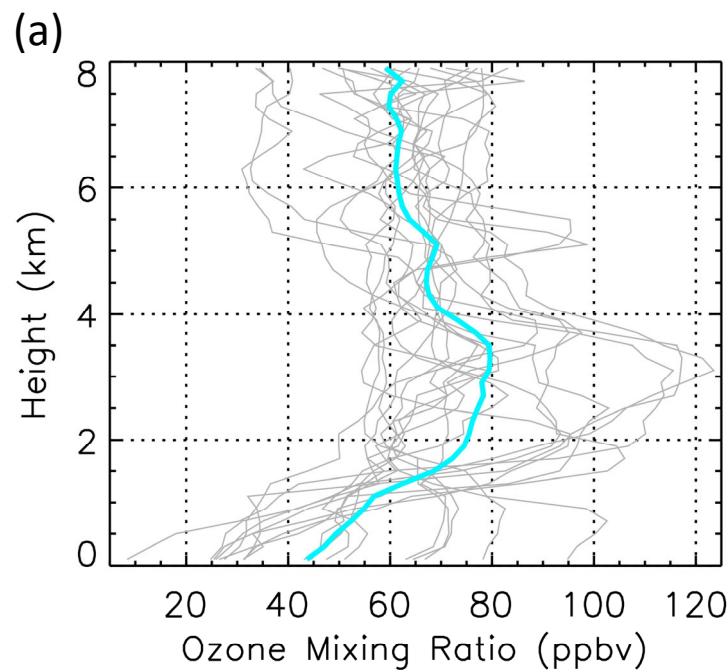


Figure 5.

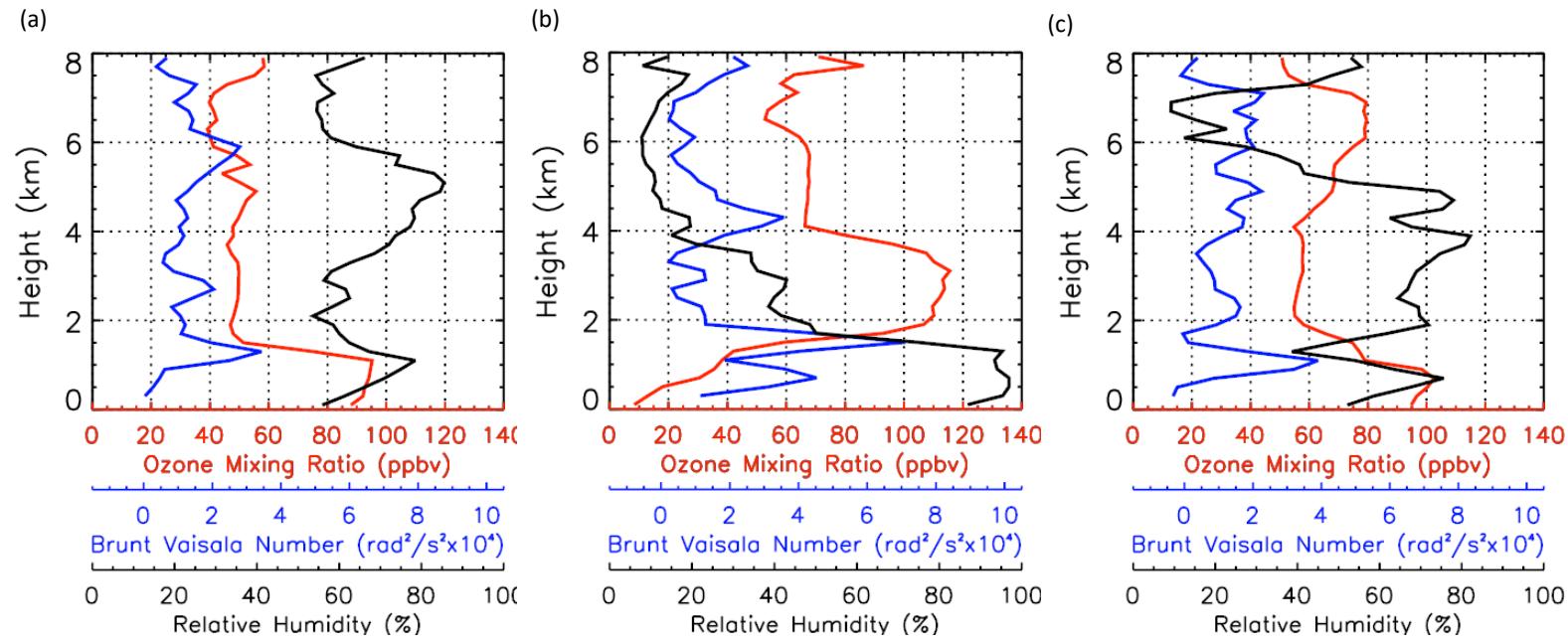


Figure 6.

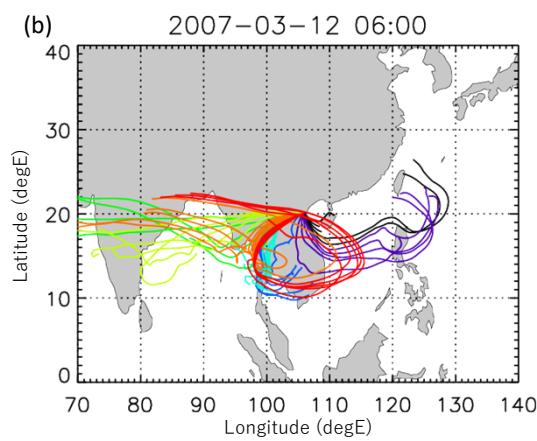
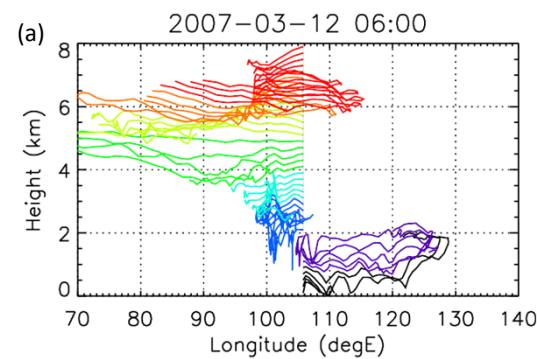


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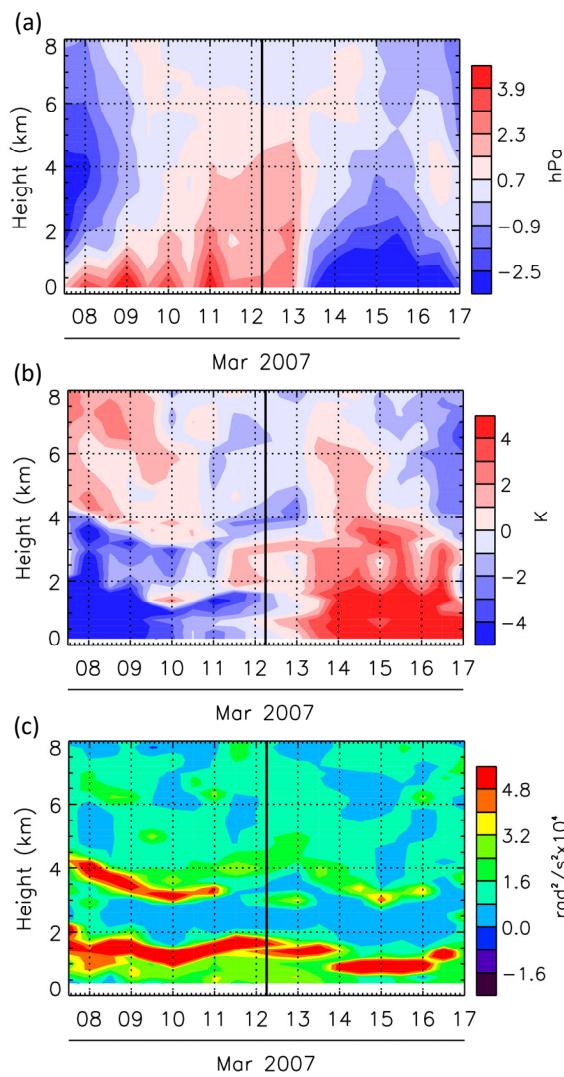


Figure 8.

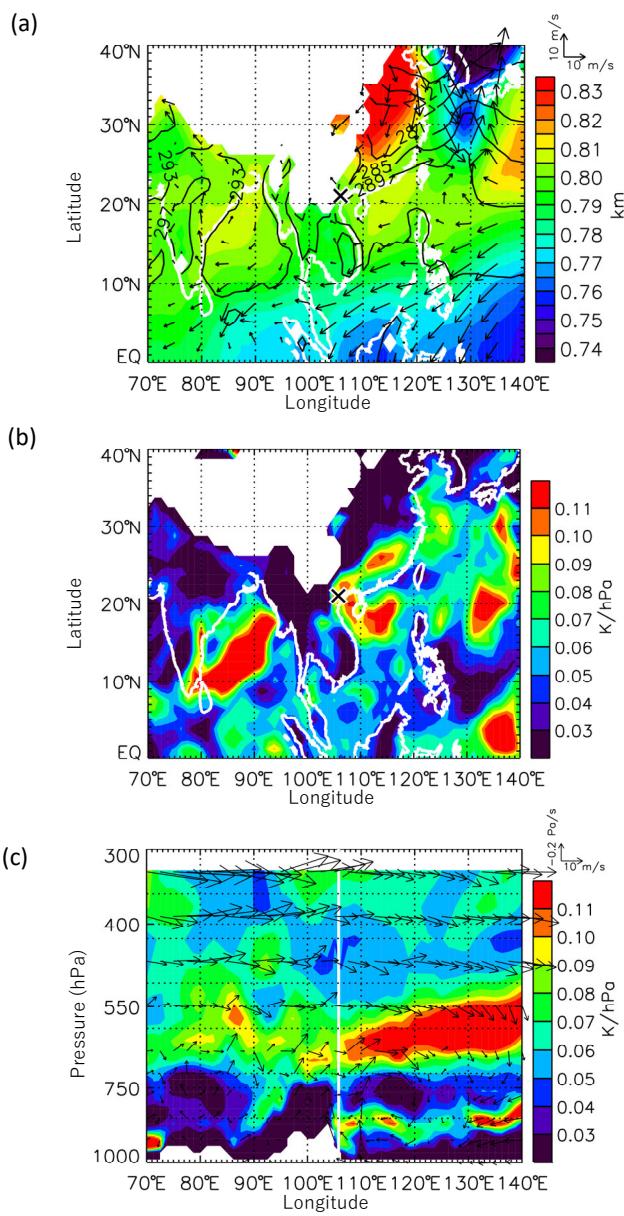


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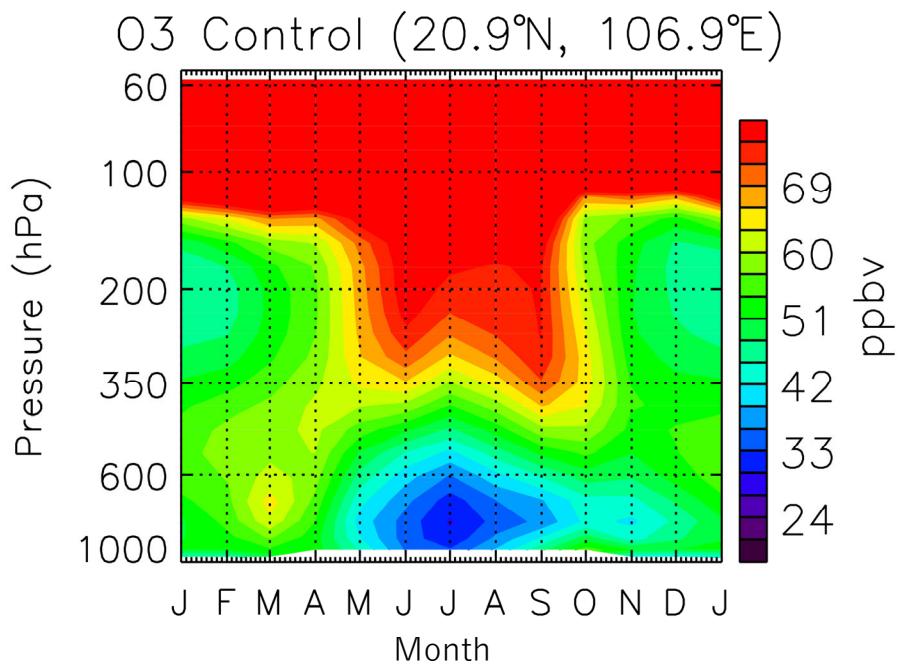


Figure 10.

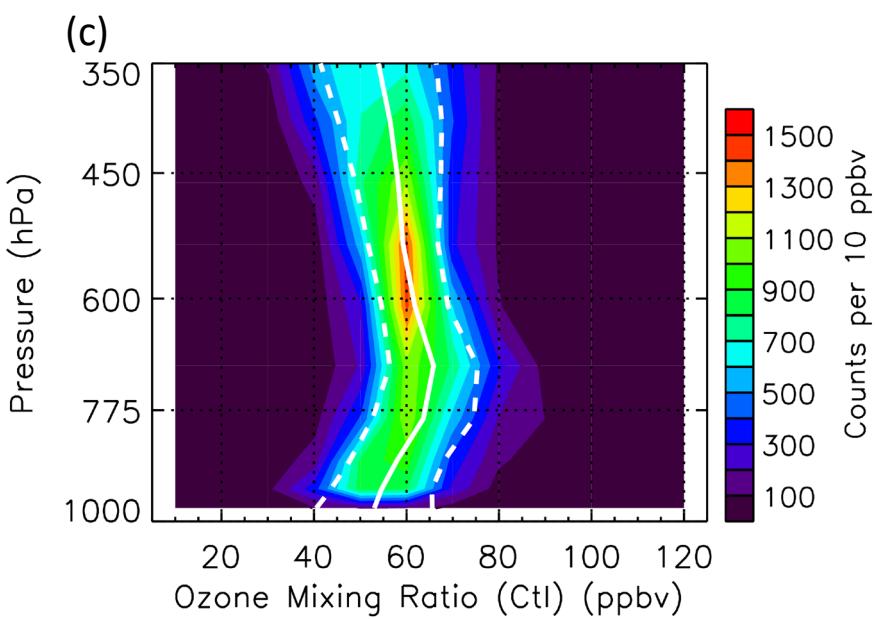
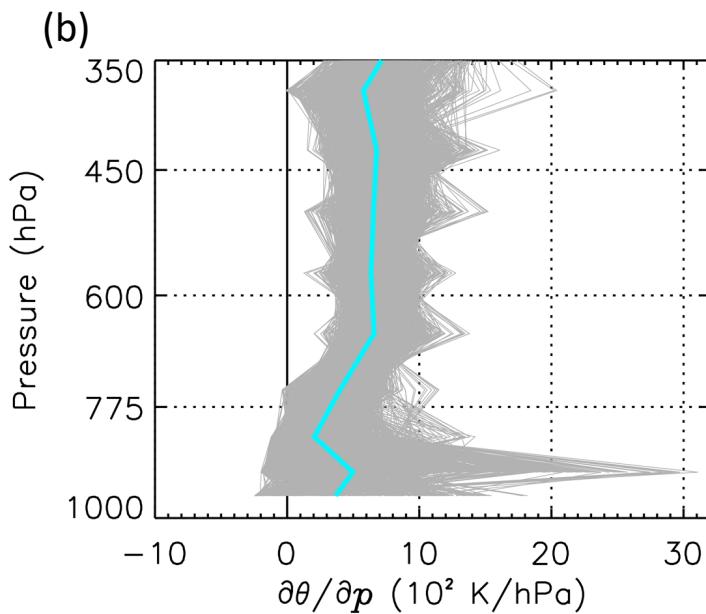
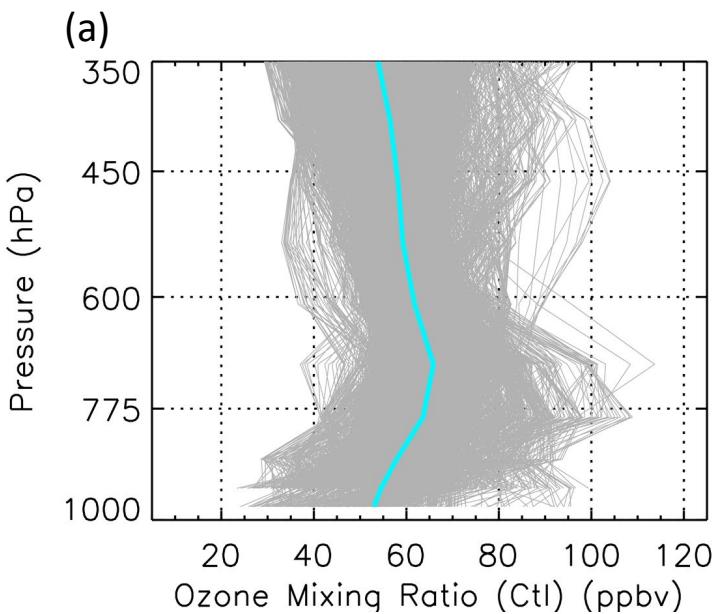


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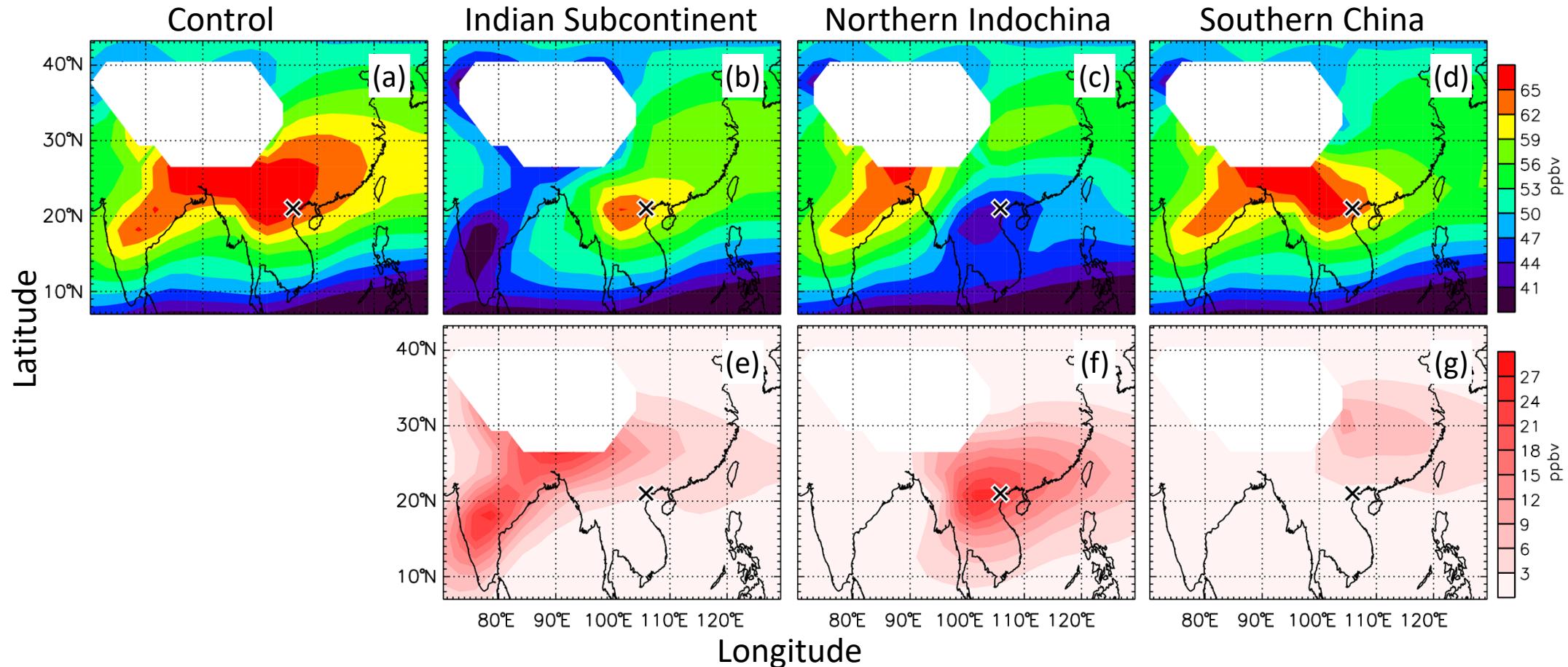


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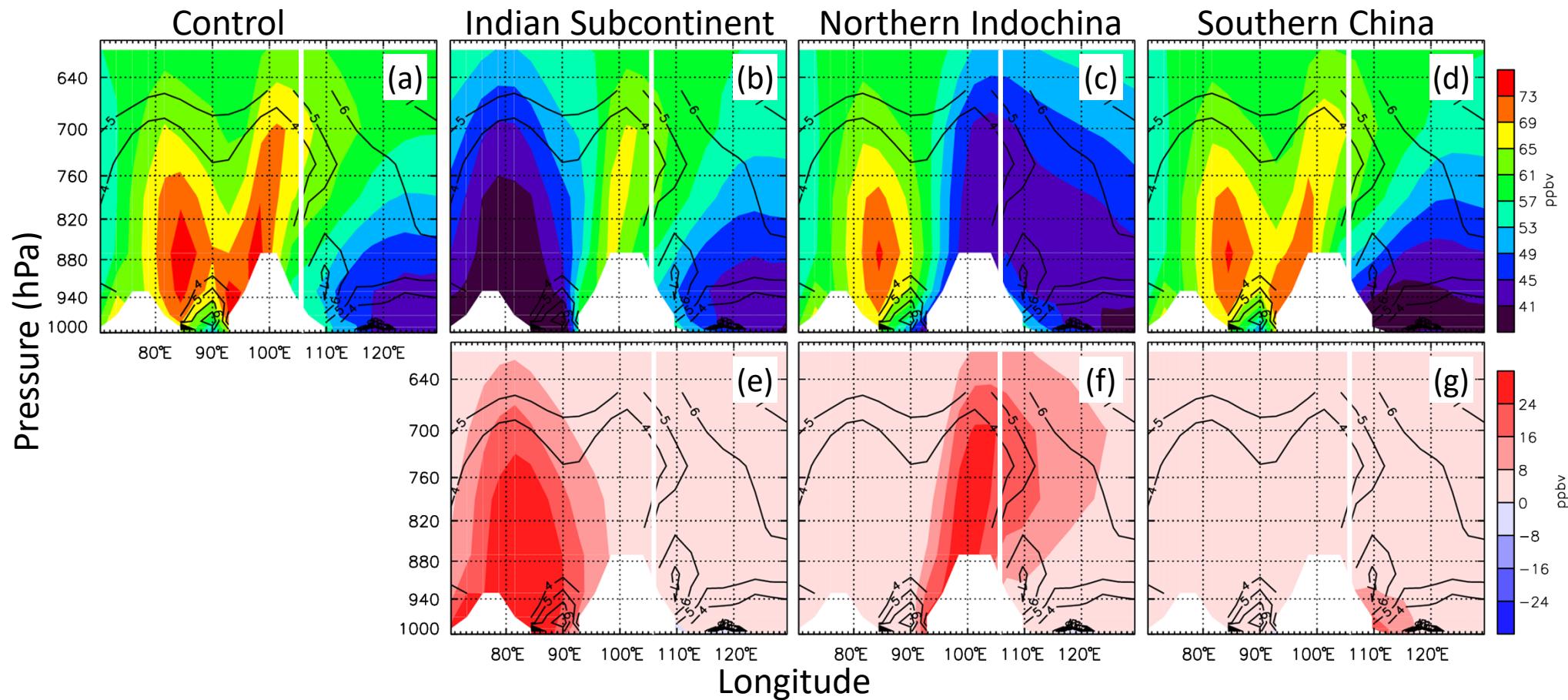


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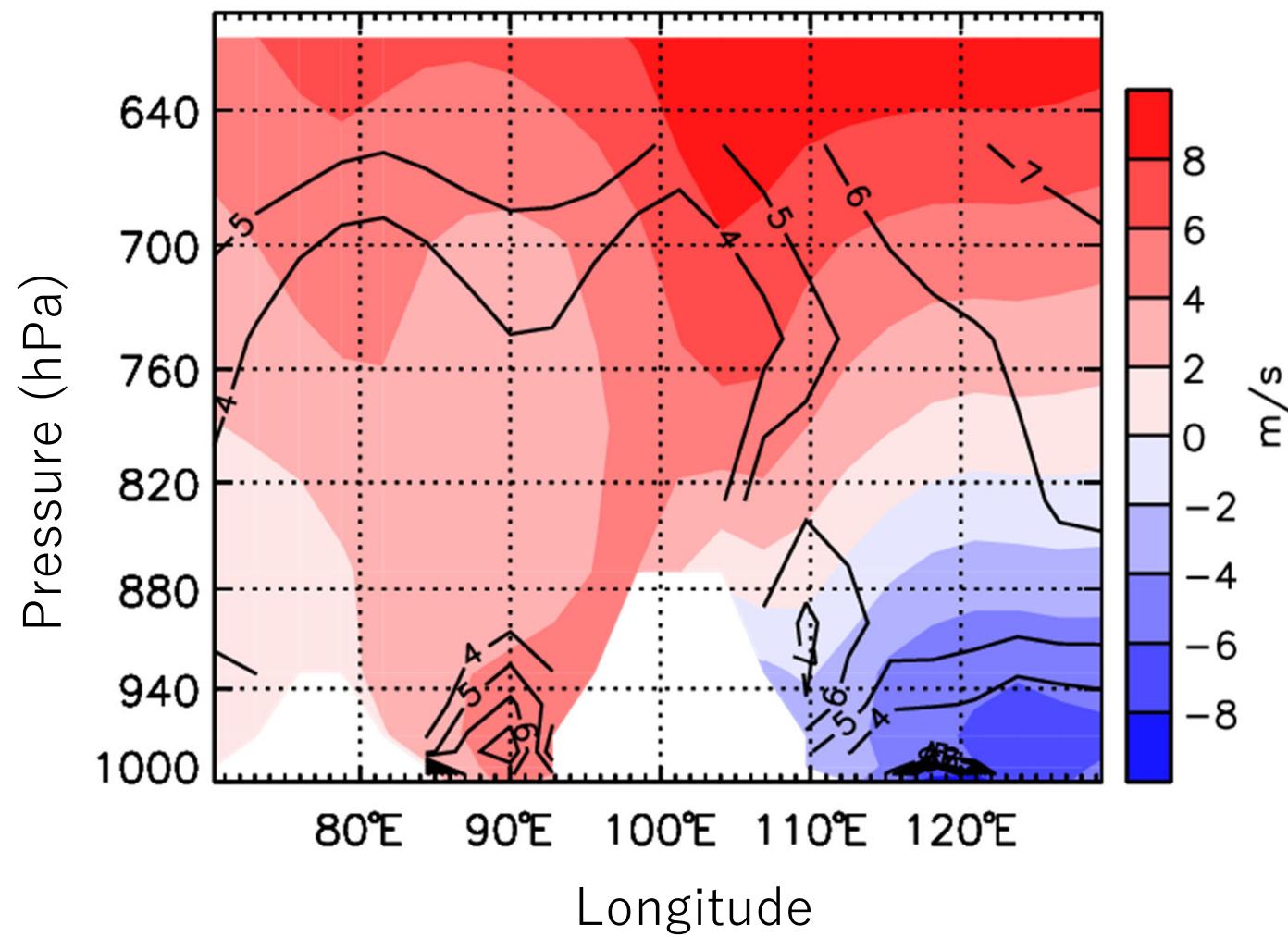


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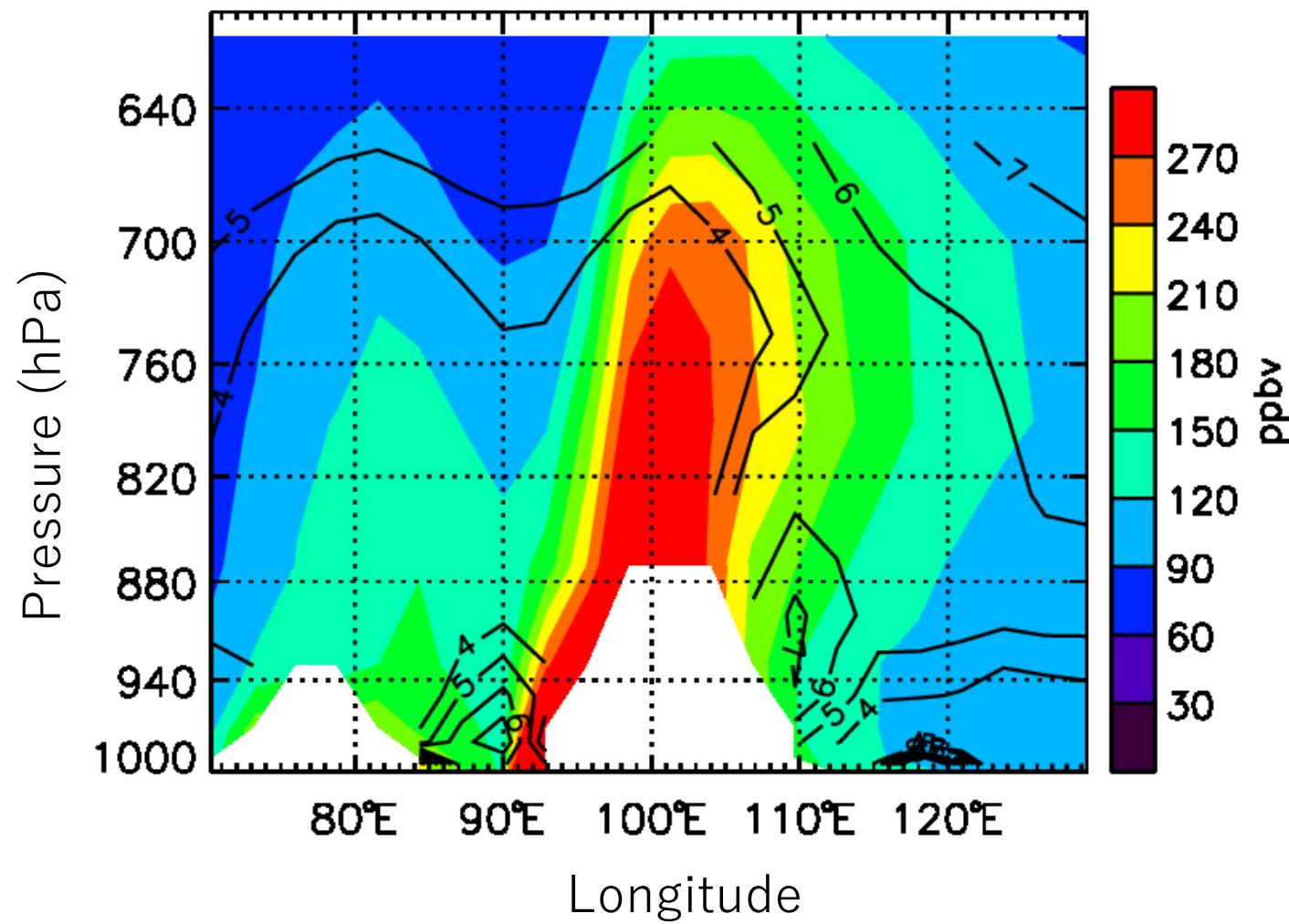


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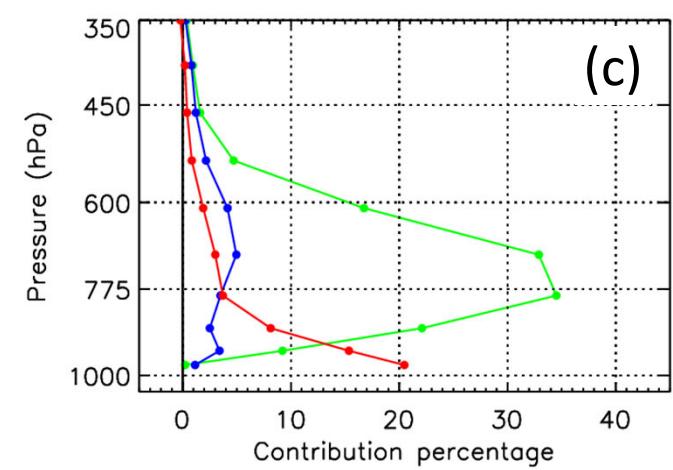
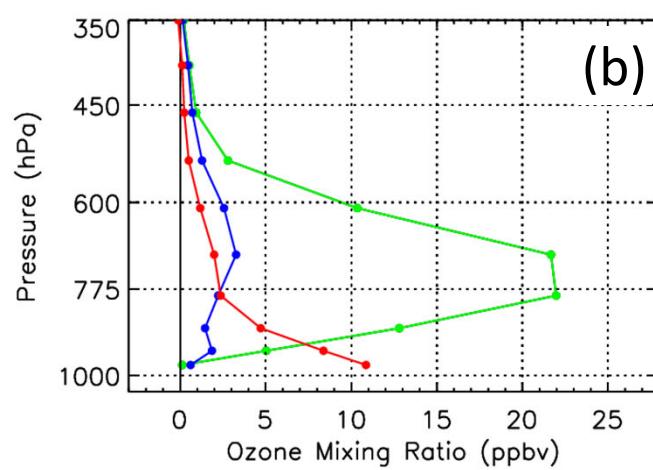
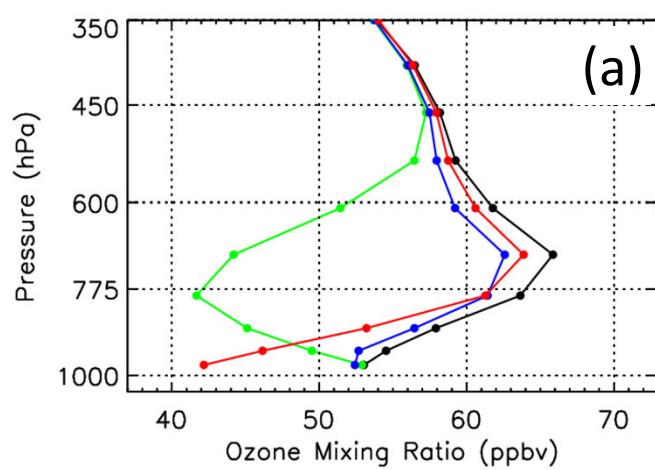


Figure 16.

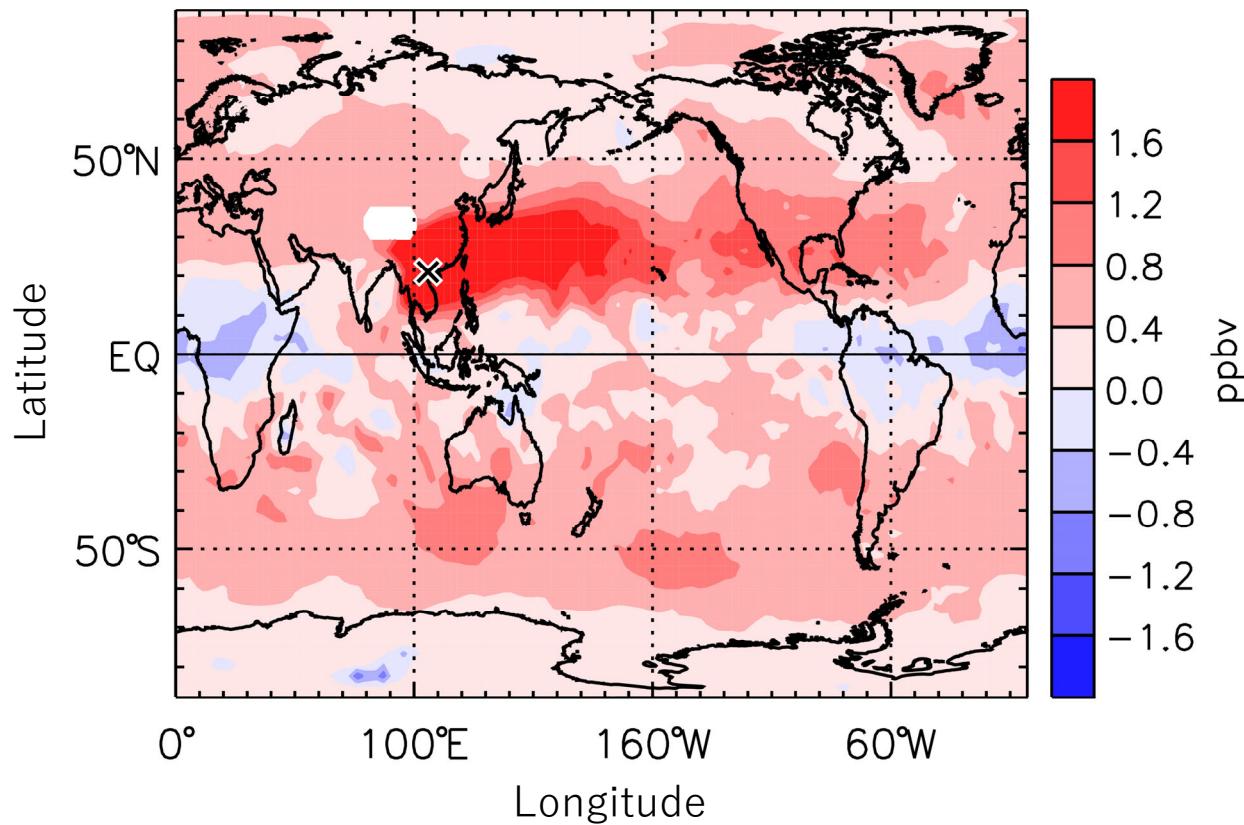


Figure 17.

